Conference or Workshop Item:
Raza, MA, Westwood, AVK orcid.org/0000-0002-5815-0429 and Stirling, C Thermal contact resistance of various carbon nanomaterial-based epoxy composites developed for thermal interface applications. In: Carbon 2018: The World Conference on Carbon, 01-06 Jul 2018, Madrid, Spain. (Unpublished)

This is an author produced version of an extended abstract presented at Carbon 2018.

THERMAL CONTACT RESISTANCE OF VARIOUS CARBON NANOMATERIAL-BASED EPOXY COMPOSITES DEVELOPED FOR THERMAL INTERFACE APPLICATIONS

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1. Introduction

Thermal interface materials (TIMs) are a vital component of electronic packaging as they facilitate heat removal from microchips by improving thermal contact between the mating surfaces of chip and heat-sink [1, 2]. Desired key characteristics of filled polymer composite TIM adhesives or pastes are high thermal conductivity, low thermal contact resistance (TCR), moderate viscosity, ease of application [3] etc. Carbon nanomaterials, such as carbon nanotubes, graphite nanoplatelets (GNPs), few layered-graphene nanosheets (FLG), vapor grown carbon nanofibers (VGCNF) and carbon black (CB) have been studied extensively for thermal interface applications due their high thermal conductivity. This work compares the heat dissipating ability by measuring TCR of epoxy composites incorporating FLG, GNPs or multiwalled carbon nanotubes (MWCNTs) under comparable conditions.

2. Materials and Methods

The epoxy resin used in this work was of a rubbery type (REP) which was developed by mixing epoxy resin and amine hardener at 1:3 (w/w) ratio [4]. Two different graphite precursors, i.e. graphite flakes and powder, were used to produce FLG and GNPs via the well-known Hummers method followed by thermal exfoliation/reduction at 900 °C for 60 s under flowing nitrogen. Commercial GNPs and MWCNTs were also used to produce composites for comparison. Composites were developed by a combined sonication and solvent mixing method (CSS). These dispersions were laid as a thin layer "bond line" between copper blocks and then cured for testing in a thermal contact resistance measurement rig, designed according to ASTM D5470 [5].
Thermal contact resistance (TCR) of nanocarbon/epoxy composites as adhesives was studied under steady state conditions following the procedures described in [5, 6]. All composites were developed at a loading of 4 wt.% for comparative analysis.

3. Results and Discussion

3.1 Thermal contact resistance of Graphene-based epoxy composites

The total TCRs of 4 wt.% FLG/REP, 4 wt.% GNP/REP, 4 wt.% MWCNT/REP composite coatings are presented in Table 1.

Table 1. The TCRs of pure REP, nanocarbon**REP composites measured at ~25 °C and under an applied pressure of 0.032 MPa at a bond line thickness of ~23 ± 5 µm.**

Composite coating	Total TCR $m^2.K/W$
4 wt.% GNP/REP produced by combined sonication and solvent mixing (CSS)	2.3×10^{-5}
4 wt.% FLG/REP by CSS	5.2×10^{-5}
4 wt.% commercial GNP/REP by CSS	5.1×10^{-5}
4 wt.% MWCNT/REP	1.36×10^{-4}
15 wt.% commercial GNP/REP produced by roll mill [7]	2.6×10^{-5}

The interfacial thermal transport performance of the MWCNT/REP composite is significantly poorer (~4.8× higher TCR at ca. 25 µm bond line thickness) than that of 4 wt.% GNP/REP

composite produced by CSS. As determined from the reciprocal gradient and y-intercept, respectively, of TCR vs. coating (i.e. bond-line) thickness plots, the latter has ~3× higher thermal conductivity and significantly lower geometric thermal contact resistance, compared to the former and these factors combine to afford the latter substantially improved performance.
In addition, the TCR of 8 wt.% MWCNT/REP adhesive is 2.4× higher than the commercial TIM (EPM 2490) at equivalent thickness of 95 μm, suggesting superior interfacial thermal transport performance for the latter. Comparisons also clearly suggest that MWCNTs are not effective fillers for production of thermal interface adhesives, compared to GNPs or FLG.

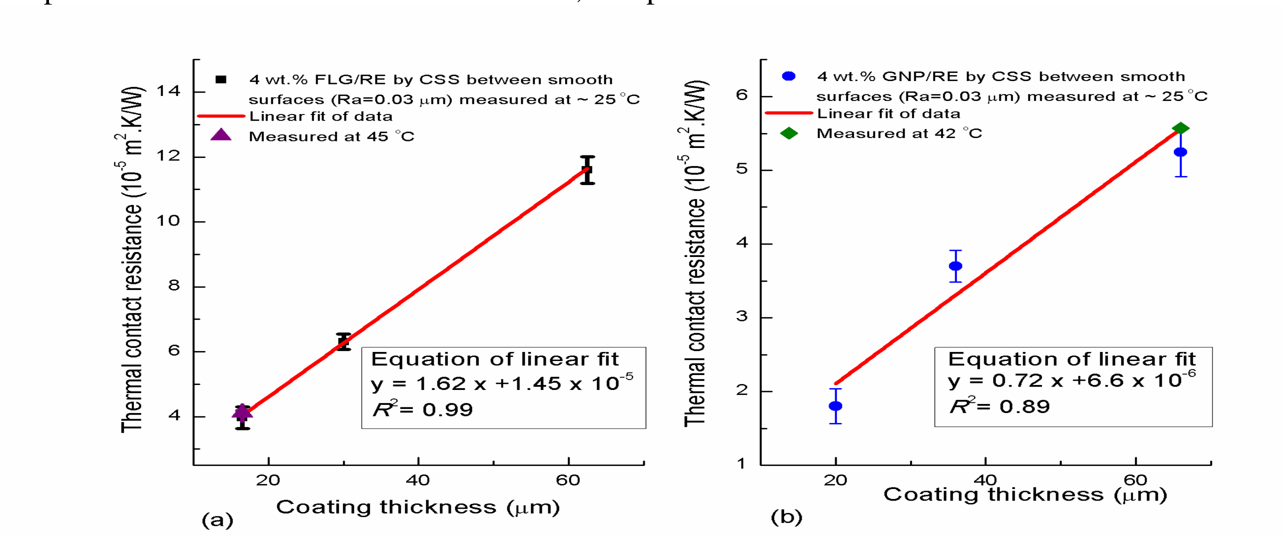

Fig. 1. Total TCR vs. coating thickness of (a) 4 wt.% FLG/REP and (b) 4 wt.% GNP/REP composites produced by combined sonication and solvent method (CSS) measured between smooth surfaces at ~0.032 MPa pressure and ~ 25 °C.

4. Conclusions

Composites produced with GNPs synthesized via Hummers' method have superior performance as TIMs compared to the composites produced with commercial GNPs at similar loading of 4 wt.%. The 4 wt.% GNP/REP composite has higher thermal conductivity and lower TCR than the corresponding composite produced with FLGs (as shown by the plots in Fig. 1) as well as vs. commercial GNP. This is attributed to the in-house GNPs being much flatter with fewer functional groups on their surface compared to the FLG and commercial GNPs. In addition, the interfacial thermal transport performance of MWCNT/REP composite was inferior to that of GNP- and FLG-based epoxy composites. This might be due to inability of MWCNT to align perpendicularly along the direction of heat flow.

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